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(54) Title: IMPROVED PEPPERMINT FLAVOR FOR ORAL HYGIENE PRODUCTS

(57) Abstract

An improved liquid oral hygiene composition containing an especially rectified flavor oil such as peppermint oil to achieve a clean, less bitter taste as well as lowering the need for the presence of menthol and surfactants. In particular, the special rectification treated peppermint oils have an advantageous effect on the efficacy and flavor of oral hygiene products containing lower amounts of alcohol.

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IMPROVED PEPPERMINT FLAVOR FOR ORAL HYGIENE PRODUCTS

FIELD OF THE INVENTION

This invention relates to oral hygiene products such as mouthwashes with an improved flavoring oil, in particular, with heavily rectified peppermint oil which is free of undesirable fractions so as to effect an uncharacteristically clean mint taste.

BACKGROUND OF THE INVENTION

The use of essential oils exhibiting penetrating characteristic flavors are well known in cosmetic, hygienic and pharmaceutical as well as food products. Natural flavor oils are generally used in the form of products of a single distillation without further processing. For example, this kind of basic flavorant product has been quite suitable for use in gums and confections. Since natural flavor oil products are poorly soluble in water, conventional liquid oral hygiene products utilize relatively high concentrations of alcohol for aiding in the solubilization and taste perception of such flavorant essential oils. Moreover, the natural extracts of essential oils contain terpenes, including sesquiterpenes. These terpene components of natural flavor oils are undesirable since they tend to oxidize upon exposure to air resulting in the development of unpleasant flavors and odors, thus limiting the useful amounts of essential oils.

The current trend to much lower alcohol content (less than 30% ethyl alcohol) in liquid oral

hygiene products, such as mouthwashes, has led to a search for reestablishing their original characteristic flavors. Consequently, the reduced alcohol content in liquid oral hygiene products resulted in a somewhat hazy appearance of the solution. This unattractive turbidity was at least partly due to coacervation, aggregation or precipitation of hydrophobic natural flavor oil components, such as terpenes. Consequently, greater amounts of surfactant substances had to be added in order to overcome the poor solubility of flavorants. At the increased concentrations, the surfactant substances would interact more frequently with the flavorants thus diminish the perceived flavor intensity of the oral hygiene products. It is well known in the art that surfactants solutions as e.g. Tween®, are apt to denature during storage. It is, therefore, desirable to apply the lowest feasible concentration of surfactant additives in liquid oral hygiene compositions so as to diminish precipitation of the components and thus extend composition shelf-life. Generally, it was also found that the decreased alcohol content of oral hygiene products also necessitated increased amounts of pleasant flavor oils. Moreover, any increased natural flavor oil concentration in turn raised the distracting impact of terpenes on the taste of these flavor (essential) oils and, in particular, mint oils. Therefore, an important goal was the removal of terpenes.

U.S. Patent No. 3,867,262 to Rockland et al., the disclosure of which is incorporated herein by reference, discloses a process for the preparation of terpeneless essential oils whereby the oil is first distilled to remove non-volatile materials. The volatile components are adsorbed onto a solid alumina adsorbent which has been pre-treated with ethyl acetate thus altering the relative affinity of the adsorbent for the terpenes and for the oxygenated components of the oil. The terpenes can be separated from the more desirable flavor constituents of the oil by elution of the adsorbent with a so-called terpenophilic solvent. An oxygenated solvent such as ethanol can be utilized to remove the oxygenated constituents which comprise the terpeneless essential oil. The Barcelon et al. U.S. Patent No. 5,030,459, discloses an improved, reduced calorie, high base content chewing gum composition containing flavor oils which are heavily rectified or redistilled by various methods.

It is, therefore, an object of the present invention to provide liquid oral hygiene compositions having low or no alcohol content which exhibit a clear solution and clean tasting flavor.

It is therefore the object of the present invention to provide liquid oral hygiene compositions containing heavily and specifically rectified oils free of undesirable fractions.

It is the further object of the present invention to provide liquid oral hygiene compositions containing heavily and specifically rectified mint oils devoid of hydrophobic, oxidizable components, such as terpenes.

It is the further object of the present invention to provide liquid oral hygiene compositions having low or no alcohol content containing heavily and specifically rectified peppermint oils permitting the use of less surfactant.

SUMMARY OF THE INVENTION

It has now been discovered that heavy and specific rectification of essential or flavoring oils not only removes undesirable fractions and contaminants but also permits a more accurate delivery of desired flavorant with an unexpectedly clean taste effect.

In particular, the present invention is directed to an improved liquid oral hygiene composition which contains heavily rectified mint flavor oils producing a less bitter clean taste and a perception of acceptable clarity thus lowering the need for surfactant agents.

The present invention utilizes a rectification or fractionation process that does not require adsorption or elution of the essential oils. The fractionation removes the "heads" and "tails" of the oils which are primarily comprised of monoterpenes and

sesquiterpenes. These are removed by vacuum distillation or on a spinning band column. By removing the monoterpenes and sesquiterpenes in this manner, the mint oils are also rendered more hydrophilic resulting in superior flavor perception attributes and lowering the requirement of alcohol in the oral vehicle.

In addition, rectification removes undesirable fractions from the oil containing resins. Rectification or redistillation of flavorants or essential oils is applied to varying degrees depending on end use. It may vary from light to heavy. Light redistillation of the oils is required to meet USP or FCC specifications. This kind of product is, of course, also suitable for gums and confections. Heavy to intermediate rectifications of flavor oils are used for oral hygiene preparations, liqueurs, boiled and high quality confections.

Accordingly, the present invention is particularly directed to a liquid oral hygiene composition with an improved minty flavor comprising (a) a flavor oil substantially free of undesirable components; (b) one or more of a surfactant composition ranging by weight from about 0.01% to about 5%; and (c) not more than about 35% by weight ethyl alcohol.

The present invention, therefore, is directed to the liquid oral hygiene composition wherein the undesirable components comprising terpenes have been

removed from the flavor oil by extraction or fractionation.

The present invention, therefore, is directed to the liquid oral hygiene composition wherein the terpenes are characterized by fractions of monoterpenes and sesquiterpenes.

The present invention is, moreover, directed to the liquid oral hygiene composition wherein the flavor oil is a mint oil.

Accordingly, the present invention is directed to the liquid oral hygiene composition wherein the mint oil is is derived from peppermint, spearmint, corn mint and mixtures thereof.

In particular, the liquid oral hygiene composition of the present invention wherein the mint oil is present in concentrations ranging from about 0.01% to about 5.0% by weight of the composition.

Specifically, a liquid oral hygiene composition is directed to the present invention having a solution of a clarity of 5 National Turbidity Units (NTU) or less or, preferably, 3 NTU or less.

An embodiment of the liquid oral hygiene composition according to the present invention further includes an intense sweetening agent the concentration ranging up to about 3.0% by weight of the composition. Further to the embodiment, the intense sweetening agent is selected from the group consisting of sodium or calcium saccharin salts, the free acid form of

saccharin, cyclamate salts, chlorosucrose derivatives, dipeptide compounds, acesulfame-K, and blends thereof.

An improved mint flavored clear mouthwash composition containing substantially terpene free peppermint oil, one or more surfactant components ranging from about 0.01% to about 3% by weight, ethyl alcohol ranging up to about 35% by weight, a sweetening agent ranging up to 3.0% by weight, an antimicrobial quaternary ammonium salt ranging from about 0.005% to about 5%, a fluorine providing compound ranging from about 0.001% to about 0.1%, a coloring agent, and a pharmaceutically acceptable buffer ranging from about 0.05% to about 0.5% by weight.

DETAILED DESCRIPTION OF THE INVENTION

"Oral composition" as defined herein pertains to a product (final or intermediate) which is not necessarily intended to be ingested but to be retained in the oral cavity, especially near or on the teeth's surfaces for a time sufficient for full contact and oral activity. "Oral vehicle" as defined herein pertains to food grade ingestible or pharmaceutically acceptable material, suitable for applying the inventive composition in the oral cavity.

Essential oils are volatile oils or essences derived from plants and usually carry the odor or flavor of the plant obtained by distillation, expression or extraction. The essential flavorant oils of the present

invention include, but are not limited to, thymol, spearmint oil, peppermint oil, lemon oil, orange oil, sage oil, rosemary oil, cinnamon oil, pimento oil, laurel oil, wintergreen oil, clove oil, and eucalyptus oil. The clarity of appearance and cleaner taste of rectified essential oils and, in particular, mint oils in oral or dental hygiene compositions according to the present invention are discussed in greater detail below.

In one preferred dental hygiene embodiment of the invention, the oral composition includes an oral vehicle and is in the form of a liquid such as a mouthwash, rinse, oral spray or other dentrifice. The concentration of the essential oil(s) ranges by weight from about 0.01% to about 1.0%.

Typical non-toxic oral carrier vehicles known in the field of dental care and hygiene may be used in the present invention. Preferred oral vehicles include water and water-alcohol mixtures. The water-alcohol mixtures are generally employed in a weight ratio from about 1:1 to about 20:1, preferably from about 3:1 to about 20:1, and most preferably from about 3:1 to about 10:1, respectively. The pH value of the vehicle is generally from about 3.0 to about 8.5, and preferably from about 4 to about 4.5. A vehicle having a pH value below about 3 is generally found as irritating to the oral cavity. A vehicle having a pH value greater than about 8.5 generally results in an unpleasant mouth feel.

The preferred embodiments of liquid oral compositions, such as mouthwashes, rinses and other dentifrice of the present invention may contain conventional additives normally employed in liquid oral hygiene compositions. These additives include sorbitol solution, a surfactant, a fluorine providing compound, a sweetening agent, a flavoring agent, a coloring agent, a humectant, a buffer, and the like, providing the additives do not interfere with the oral hygiene properties of the composition of the present invention.

The liquid oral clinically effective oral hygiene compositions of the present invention may contain sorbitol or sorbitol solution in high weight to volume concentrations (w/v), i.e., from about 10% to about 60%, and preferably from about 15% to about 40%, of sorbitol solution, U.S. Pharmacopeia, which is a solution of sorbitol in water containing 70% total solids. Sorbitol solution supplies sweetness and body to the composition and gives a desirable mouth feel. Sorbitol solution also enhances flavor, prevents harsh taste and provides a fresh and lively sensation in the mouth.

Surfactants (surface active agents) are organic compounds which reduce surface tension between liquids and aid in the dispersion of a composition throughout the oral cavity. The surfactant in the present invention may be nonionic, ampholytic, or cationic. The liquid oral hygiene compositions of the

present invention may contain surfactants in amounts up to about 5%, preferably from about 0.01% to about 3%, and more preferably from about 0.05% to about 2%, by weight of the liquid oral hygiene composition.

Suitable nonionic surfactants in the present invention include poly(oxyethylene)-poly(oxypropylene) block copolymers. Such copolymers are known commercially by the non-proprietary name of poloxamers, which name is used in conjunction with a numeric suffix to designate the individual identification of each copolymer. Poloxamers may have varying contents of ethylene oxide and propylene oxide which results in poloxamers which have a wide range of chemical structures and molecular weights. The nonionic poloxamer surfactants of the present invention are non-toxic, are acceptable as direct food additives, are stable, readily dispersible in aqueous systems and are compatible with the wide variety of formulating ingredients used in oral compositions.

Poloxamer surfactants in the present invention should have a Hydrophilic-Lipophilic Balance (HLB) of between about 10 and about 30, and preferably between about 10 and about 25. Suitable poloxamers in this invention include: Poloxamers 105, 108, 123, 124, 183, 184, 185, 188, 215, 217, 234, 235, 237, 238, 284, 288, 334, 335, 338, and 407. A particularly preferred poloxamer is Poloxamer 407, which has an HLB of about 22, and is sold under the tradename Pluronic F-127 by

BASF Wyandotte, Parsippany, N.J. When present in the liquid oral hygiene composition, poloxamers should constitute from about 0.01% to about 5%, and preferably from about 0.05% to about 1%, by weight of the total volume of liquid oral hygiene composition (w/v).

Another class of nonionic surfactants useful in the oral composition of this invention are the ethoxylated hydrogenated castor oils. These surfactants are prepared by hydrogenating castor oil and treating the hydrogenated product with from about 10 to about 200 moles of ethylene glycol. These ethoxylated hydrogenated castor oils are known by the non-proprietary name of PEG hydrogenated castor oils, in accordance with the dictionary of the Cosmetics, Toiletries and Fragrance Association, 3rd Edition, which name is used in conjunction with a numeric suffix to designate the degree of ethoxylation of the hydrogenated castor oil product, i.e., the number of moles of ethylene oxide added to the hydrogenated castor oil product. Suitable PEG hydrogenated castor oils include PEG 16, 20, 25, 30, 40, 50, 60, 80, 100, and 200. In a preferred embodiment of the oral compositions, the PEG hydrogenated castor oil surfactant is Cremophor RH40, a commercially available product from BASF-Wyandotte, Parsippany, N.J. Ethoxylated hydrogenated castor oil surfactants, when present in the liquid oral dental hygiene composition for combatting plaque and gingivitis, should constitute from about 0.2% to about

2%, and preferably from about 0.5% to about 1%, by weight of the total volume of liquid oral hygiene composition (w/v).

Other nonionic surfactants useful in the present invention include condensates of sorbitan esters of fatty acids with ethylene oxide (polysorbates) such as sorbitan mono-oleate with from about 20 to about 60 mole percent of ethylene oxide (e.g., "Tween", a trademark of ICI U.S., Inc.) Particularly preferred polysorbates are Polysorbate 20 (polyoxyethylene 20 sorbitan monolaurate, Tween 20) and Polysorbate 80 (polyoxyethylene 20 sorbitan mono-oleate, Tween 80).

Additional suitable nonionic surfactants useful in the oral composition of the present invention are the condensation products of an alpha-olefin oxide containing 10 to 20 carbon atoms, a polyhydric alcohol containing 2 to 10 carbon atoms and 2 to 6 hydroxyl groups, and either ethylene oxide or a mixture of ethylene oxide and propylene oxide. The resultant surfactants are polymers which have a molecular weight in the range from about 400 to about 1600, contain from about 40% to about 80% ethylene oxide, by weight, and have an alpha-olefin oxide to polyhydric alcohol mole ratio in the range from about 1:1 to about 1:3, respectively.

Other nonionic surfactants useful in the present invention include condensates of sorbitan esters of fatty acids with polyethylene glycol such as sorbitan

diisostearate condensed with polyethylene glycol. A particularly preferred polyethylene glycol condensate of a diisostearate sorbate ester is EmSorb 2726, a commercially available product manufactured by Emery Industries Incorporated, Linden, N.J.

Amphoteric surfactants have the capacity to behave as either an acid or a base. Amphoteric surfactants useful in the present invention include quaternized imidazole derivatives.

Cationic surfactants are surfactants which carry a positive charge. Cationic surfactants especially useful in the present invention include antimicrobial quaternary ammonium salts. This class of compounds can be illustrated but not limited to, cetylpyridinium chloride, benzalkonium chloride, benzethonium chloride, domiphen bromide, 1-(3-chlorallyl)-3,5-7-triaza-1-azoniaadamantane chloride, and menthene ammonium chloride. A preferred embodiment of the present invention provides antimicrobial quaternary ammonium salt detergent ranging from about 0.005% to about 5%.

Fluorine providing compounds may be fully or slightly water soluble and are characterized by their ability to release fluoride ions or fluoride containing ions in water and by their lack of reaction with other components in the composition. Typical fluorine providing compounds are inorganic fluoride salts such as water-soluble alkali metal, alkaline earth metal, and

heavy metal salts, for example, sodium fluoride, potassium fluoride, ammonium fluoride, cuprous fluoride, zinc fluoride, stannic fluoride, stannous fluoride, barium fluoride, sodium fluorosilicate, ammonium fluorosilicate, sodium fluorozirconate, sodium monofluorophosphate, aluminum mono- and difluorophosphates and fluorinated sodium calcium pyrophosphate. Alkali metal fluorides, tin fluoride and monofluorophosphates, such as sodium and stannous fluoride, sodium monofluorophosphate and mixtures thereof, are preferred.

The amount of fluorine providing compound present in a preferred embodiment of the liquid oral hygiene composition is dependent upon the type of fluorine providing compound employed, the solubility of the fluorine compound, and the nature of the final liquid oral hygiene composition. The amount of fluorine providing compound used must be a nontoxic amount. In general, the fluorine providing compound when used will be present in an amount up to about 1%, preferably from about 0.001% to about 0.1%, and most preferably from about 0.001% to about 0.05%, by weight of the liquid oral hygiene composition.

When sweetening agents (sweeteners) are used, those sweeteners well-known in the art, including both natural and artificial sweeteners, may be employed. The sweetening agent used may be selected from a wide range of materials including water-soluble sweetening agents,

water-soluble artificial sweetening agents, water-soluble sweetening agents derived from naturally occurring water-soluble sweetening agents, dipeptide based sweetening agents, and protein based sweetening agents, including mixtures thereof. Without being limited to particular sweetening agents, representative categories and examples include:

(a) water-soluble sweetening agents such as monosaccharides, disaccharides and polysaccharides such as xylose, ribose, glucose (dextrose), mannose, galactose, fructose (levulose), sucrose (sugar), maltose, invert sugar (a mixture of fructose and glucose derived from sucrose), partially hydrolyzed starch, corn syrup solids, dihydrochalcones, monellin, steviosides, glycyrrhizin, and sugar alcohols such as sorbitol, mannitol, maltitol, hydrogenated starch hydrolysates and mixtures thereof;

(b) water-soluble artificial sweeteners such as soluble saccharin salts, i.e., sodium or calcium saccharin salts, cyclamate salts, the sodium, ammonium or calcium salt of 3,4-dihydro-6-methyl - 1,2,3-oxathiazine-4-one-2, 2-dioxide, the potassium salt of 3,4-dihydro-6-methyl - 1,2,3-oxathiazine-4-one-2, 2-dioxide (Acesulfame-K), the free acid form of saccharin, and the like;

(c) dipeptide based sweeteners, such as L-aspartic acid derived sweeteners, such as L-aspartyl-L-phenylalanine methyl ester (Aspartame) and

materials described in U.S. Pat. No. 3,492,131, L-alphaaspartyl-N-(2,2,4,4-tetramethyl-3-thietanyl)-D-alaninamide hydrate (Alitame), methyl esters of L-aspartyl-L-phenylglycerine and L-aspartyl-L-2,5-dihydrophenylglycine, L-aspartyl-2,5-dihydro-L-phenylalanine; L-aspartyl-L-(1-cyclohexen)-alanine, and the like;

(d) water-soluble sweeteners derived from naturally occurring water-soluble sweeteners, such as chlorinated derivatives of ordinary sugar (sucrose), known, for example, under the product designation of Sucralose; and

(e) protein based sweeteners such as thaumaoccous danielli (Thaumatin I and II).

In general, an effective amount of sweetening agent is utilized to provide the level of sweetness desired in any particular liquid oral hygiene compositions, and this amount will vary with the sweetener selected and the final liquid oral hygiene product. The amount of sweetener normally present is in the range from about 0.0025% to about 90%, by weight of the embodiment of the liquid oral hygiene composition such as mouthwash or rinse, depending upon the sweetener used. The exact range of amounts for each type of sweetener is well known in the art and is not the subject of the present invention. The flavoring agents (flavors, flavorants) which may be used include those flavors known to the skilled artisan, such as natural

and artificial flavors. Suitable flavoring agents include mints, such as peppermint, citrus flavors such as orange and lemon, artificial vanilla, cinnamon, various fruit flavors, both individual and mixed and the like.

The amount of flavoring agent employed herein is normally a matter of preference subject to such factors as the type of final liquid oral hygiene composition, the individual flavor and the strength of flavor desired. Thus, the amount of flavoring may be varied in order to obtain the result desired in the final product and such variations are within the capabilities of those skilled in the art without the need for undue experimentation. The flavoring agents, when used, are generally utilized in amounts that may, for example, range in amounts from about 0.05% to about 6%, by weight of the liquid oral hygiene composition.

The coloring agents (colors, colorants) useful in the present invention are used in amounts effective to produce the desired color. The colorants may include natural food colors and dyes suitable for food, drug and cosmetic applications. These colorants are known as F. D. & C dyes and lakes. The materials acceptable for the foregoing uses are preferably water-soluble. Illustrative nonlimiting examples include the indigoid dye known as F. D. & C Blue No.2, which is the disodium salt of 5,5-indigotindisulfonic acid. Similarly, the dye known as F. D. & C Green No.1 comprises a

triphenylmethane dye and is the monosodium salt of 4-[4-(N-ethyl-p-sulfoniumbenzylamino) diphenylmethylene]-[1-(N-ethyl-N-p-sulfoniumbenzyl)-delta-2,5- cyclohexadieneimine]. A full recitation of all F.D.& C colorants and their corresponding chemical structures may be found in the Kirk-Othmer Encyclopedia of Chemical Technology, 3rd Edition, in volume 5 at pages 857-884, which text is incorporated herein by reference.

Suitable humectants in the oral composition of the present invention include glycerin, propylene glycol, polyethylene glycol, sorbitan, fructose, mixtures thereof and the like. Humectants, when employed, may be present in amounts from about 10% to about 60%, by weight of the liquid oral hygiene composition.

Suitable buffers in the present invention include citric acid-sodium citrate, phosphoric acid-sodium phosphate, acetic acid-sodium acetate and benzoic acid and benzoate in amounts up to about 1%, and preferably from about 0.05% to about 0.5%, by weight of the liquid oral hygiene composition for dental hygiene.

The present invention extends to methods of making the improved liquid oral antiseptic compositions. The final oral compositions are readily prepared using methods generally known by those skilled in the dental art. In such a method, an oral antiseptic composition,

according to the present invention is made by first dissolving the surfactant, cetylpyridinium chloride, in water, then admixing sorbitol or sorbitol solution to the surfactant solution until the sorbitol is dissolved. Coloring agents, additional sweetening agents, and similar additives are admixed at the same time sorbitol is added. Flavorants, such as peppermint oil, eucalyptol, and thymol, for example, are then admixed to the surfactant/sorbitol solution until dissolved. The pH value of the solution is adjusted to 4-6 using 1-N hydrochloric acid or 1-N sodium hydroxide. Then sufficient water or alcohol, or mixtures thereof are added to the solution with mixing until the final solution volume is reached. In a preferred embodiment, peppermint oil, eucalyptol, and thymol are conveniently added to the solution in the alcohol portion if any.

The apparatus useful in accordance with the present invention comprises mixing apparatus well known in the dental art, and therefore the selection of the specific apparatus will be apparent to the artisan.

Mint oils (from the Greek Mintho) are well known throughout the ages and originated in the Mediterranean Basin. Simple distillation of mint oils was traced to writings as early as 410 A.D. In fact both herb and oil of mint plants were used widely for medicinal and culinary purposes. In particular, preparations and prescriptions from mint herbs became the medicines for most ailments in medieval times.

The desirable fragrance and mild, stinging, pleasant taste are the main properties of mint. The main desirable taste ingredient of peppermint oil is menthol, and that of spearmint is carvone. In general, the oil should be free of adulterants, contaminants and chemical residues. In addition, the oil should be colorless to only slightly yellow with good odor and characteristic prime quality taste. Peppermint oils are extracted mainly from the leaf of mint plants. Its yield varies from one growing region to another. For the purpose of uniformity and consistency as well as purity or lack of contaminant toxic substances, comparative analyses and evaluation of the flavoring oils are conducted to afford overall standardization of mint oil blends. Table I shows an comparative analysis of peppermint oils from several different sources by gas chromatography. Table II shows similarly obtained chromatographic data of spearmint oil constituents from four sources, Midwest Native, Midwest Scotch, Farwest Native and Farwest Scotch.

TABLE I

PEPPERMINT OILS

	R.T	MAD	MID	WIL	YAK	EOI
Isobutyraldehyde	5.20	.03	.02	.02	.02	.02
Isovaleraldehyde	6.07	.17	.19	.15	.10	.14
2-Ethylfuran	6.42	.03	.02	.02	.02	.02
α-Pinene	7.55	.70	.79	.67	.76	.69
β-Pinene	9.46	.94	1.08	.95	1.02	.94
Sabinene	9.76	.46	.51	.49	.50	.53
Myrcene	10.71	.15	.17	.13	.19	.16
α-Terpinene	11.40	.36	.35	.40	.35	.42
L-Limonene	11.97	1.30	1.44	1.28	1.68	1.69
1,8-Cineole	12.29	5.40	6.44	5.30	5.74	5.81
Cis-Ocimene	12.83	.27	.27	.24	.32	.27
Gamma-Terpinene	13.39	.70	.66	.79	.68	.86
Para-Cymene	14.27	.15	.16	.20	.15	.16
Terpinolene	14.62	.15	.15	.16	.17	.17
3-Octanol	17.90	.16	.15	.16	.19	.16
1-Octene-3-ol	19.85	.11	.13	.10	.11	.09
T-Sabinenehydrate	20.60	1.07	.86	1.12	1.06	1.14
L-Menthone	21.14	20.44	26.98	21.69	18.03	21.43
Menthofuran	21.63	3.06	2.18	1.86	6.89	3.54
δ-Isomenthone	22.09	2.65	3.43	2.51	2.58	2.92
β-Bourbonene	22.83	.42	.20	.26	.24	.26
Linalool	23.29	.27	.26	.23	.38	.27
Menthyl acetate	24.12	4.54	2.99	4.97	4.01	3.43
Neo-menthol	25.07	3.11	3.11	3.48	2.81	3.15
Terpinene-4-ol	25.42	.98	.99	1.04	.84	1.18
β-Caryophellene	25.53	1.92	1.92	1.98	1.66	1.57
L-Menthol	26.53	45.84	41.69	47.45	43.17	44.02
Trans-β-Farnesene	27.40	.23	.18	.25	.23	.22
Pulegone	27.43	1.60	.58	.92	2.54	1.87

	R.T	MAD	MID	WIL	YAK	EOI
α-Terpineol	28.76	.37	.23	.20	.23	.19
Germacrene-D	29.80	1.57	1.62	1.48	1.47	1.15
Piperitone	30.40	.73	.63	.75	.58	.44
Viridiflorol	40.31	.16	.21	.16	.17	.15

RT : Retention time in minutes
MAD : Madras
MID : Midwest
WIL : Willamette
YAK : Yakima
EOI : Eastern Oregon/Idaho

TABLE II

SPEARMINT OILS

	R.T	MWN	FWN	MWS	FWS
Isobutyraldehyde	5.20	.02	.03	.02	.03
Isovaleraldehyde	6.07	.16	.15	.09	.11
2-Ethylfuran	6.42	.03	.03	.02	.02
α-Pinene	7.55	.66	.70	.61	.66
β-Pinene	9.46	.59	.59	.59	.57
Sabinene	9.76	.47	.51	.49	.59
Myrcene	10.71	2.04	2.41	.71	.92
α-Terpinene	11.40	.24	.31	.22	.29
Limonene	11.97	10.66	11.58	17.10	19.00
1,8-Cineole	12.29	1.95	2.13	1.64	1.39
Cis-Ocimene	13.09	.23	.22	.05	.05
Gamma-Terpinene	13.59	.56	.60	.48	.58
Para-Cymene	14.27	.06	.05	.02	.03
Terpinolene	14.84	.15	.15	.13	.13
3-Octyl acetate	16.42	.19	.15	.13	.16
3-Octanol	18.05	.59	.67	1.93	1.67
T-Sabinenehydrate	20.70	1.22	.92	.11	.12
L-Menthone	21.55	.03	.03	1.11	.31
β-Bourbonene	22.98	1.53	1.17	.86	.83
Linalool	23.58	.12	.10	.11	.09
Terpinene-4-ol	25.42	.97	1.36	.50	.76
β-Caryophellene	25.60	1.13	1.21	.90	1.44
Dihydrocarvone	26.04	.69	1.08	1.00	1.34
Dihydrocarvyl acetate	26.50	.08	.07	.03	.06
Trans-β-Farnesene	27.40	.93	.93	.19	.23
α-Terpineol	28.34	.33	.32	.43	.56
Germacrene-D	29.18	.26	.35	.20	.33
Carvone	30.06	70.40	69.67	70.78	69.27
Carvyl acetate	31.41	1.10	.77	.10	.09
Trans-Carveol	32.91	.28	.25	.24	.23

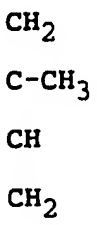
	R.T	MWN	FWN	MWS	FWS
Cis-Carveol	33.83	.25	.23	.19	.22
Cis-Jasmone	36.82	.31	.25	.11	.12
Viridiflorol	41.62	.21	.22	.02	.02

RT : Retention time in minutes
MWN : Midwest Native
FWN : Farwest Native
MWS : Midwest Scotch
FWS : Farwest Scotch

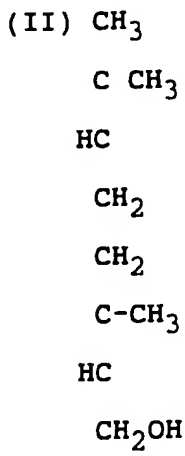
An important aspect of the present invention is the special fractionation of the oils to improve quality. This method achieves other special benefits as well as, e.g., enhanced flavor penetration or perception, enhanced solubility, and less bitterness in liquid oral hygiene compositions (mouthwashes or breath cleansers) and more minty and cleaner tasting peppermint flavor oil.

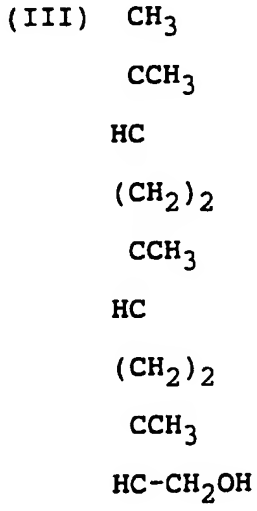
The flavor oil that is incorporated into the oral hygiene composition has been fractionated or rectified using standard distillation equipment to remove the terpene components that act as flavor detractants. This is carried out through standard distillation procedures using a vacuum distillation apparatus or a spinning band column. Terpenes themselves are simple, non-saponifiable lipids which are multiplies of the isoprene molecule (C_5H_8) whose molecular formula (I) is set forth below.

(I)



Monoterpenes (II) are basically comprised of two of the above isoprene units whereas sesquiterpenes (III) are comprised of three of these units.





Standard deterpenation, which results in removal of most of the "head" (monoterpene) fraction from the essential oil is also beneficial in providing enhanced flavor perception the fractionation of the oil is removing the "head" as well as the "tail" fractions.

An additional type of fractionation of peppermint oil involves done by extensive distillation or rectification whereby more of the head and tail as well as specific middle fractions were removed. It is evident that repeated distillations in this manner remove not only the mono- and sesquiterpenes but the menthonic fraction as well in a number of the mint flavor oils.

The major constituents of the "menthonic" fraction are menthone, its isomer iso-menthone, and menthofuran. Menthone has been traditionally understood to be an essential component of peppermint oil. Reducing the oil to more than 40% of the menthone and iso-menthone content as well as removal of the major portion of menthofuran has also resulted in an improved quality oil. The improved oil possessed a clean mentholic sensation without the heavy herbal and resinous character. The oil had a reduced bitter taste and it would appear that the removed menthonic fraction and the tail fraction seem to contribute to the unpleasant bitter taste of peppermint.

Flavoring oils of particular utility in the practice of the present invention are those of the mint variety (Genus Mentha) such as peppermint (Mentha Piperita L.), and spearmint (Mentha Spicatal and Mentha Cardiac G). Originally from the Mediterranean Basin mint plants were introduced in the Eastern region of North America during the colonial times. (Massachusetts, New York and New Jersey) moving gradually to the Midwest (Ohio, Indiana and Michigan). More recently, in the last 70 years, the plant has been grown in the Far West (Oregon, Idaho, Montana and Nevada). Corn mint (Mentha Arvensis) is grown in China, Japan and South America and not considered "peppermint" in the U.S.A.. The amount of deterpenated flavor oil employed is normally a matter of preference but it has

been found that amounts ranging from approximately 0.01% to approximately 5.0% by weight of the final composition are usable with amounts of about 0.1% to about 3% by weight being preferred. The mint oils of the present invention can be utilized as the sole flavorant or can be combined with other standard single distillation mint oils.

Procedures for additional purification of the mint oil are illustrated by the following methods. In particular, oils with different tastes and taste intensities resulted from different procedures of rectification of the peppermint oils. The cleaner and less bitter flavor of the highly rectified oils was distinctly perceptible by comparison with standard low rectification oil.

Highly rectified peppermint oils and highly rectified deterpenated peppermint oils were compared in certain preferred embodiments of the mouthwashes in accordance with the present invention. First, the highly rectified deterpenated oils were devoid of monoterpene fractions as the so-called "heads" fraction and the major portion of the so-called "tails" fraction comprising sesquiterpene compounds and derivatives thereof. Highly rectified peppermint oils which are only about 50% relieved of the head monoterpene fractions and the tail sesquiterpenes fractions.

The standard control was redistilled to conform with USP specification; the procedure entails only a minor portion of the oil.

Among the presently preferred peppermint oils are the prime quality Madras product from the Madras region in Oregon (U.S.A.), Rose Mitcham, rectified oil from a blend of Farwest Williamette, Midwest and Madras, and Farwest Terpeneless VR, a heavy fractionation of a blend of prime quality natural peppermint oils from the Madras Williamette Farwest producing regions of the U.S.A. This product is steam distilled from fresh overground parts of the flowering plant, Mentha Piperata L.

The Farwest terpeneless oil is produced by the procedure essential oils, as e.g., similar to the rectification of lemon oil disclosed in U.S. Patent 3,867,262 as incorporated by reference, for illustration purposes:

A. Molecular Distillation to Separate Non-volatile Matter

Twenty-five ml. of commercial, raw, cold-pressed Desert-type lemon oil was molecularly distilled in a Kontes D-285600 falling film molecular still operated at 10 Torr during a 6-hour period. The volatile components were collected as follows: 35% in the distillate receiver and 65% in the Dry-ice trap between the high vacuum pump and the distillate

receiver. These two fractions were combined (total volume 23.5 ml.) for use in Part C.

The distillation residue (0.9 g.) consisted largely of high boiling waxes, coumarins, and unidentified solids and contained only insignificant traces of volatile terpene hydrocarbons and oxygenated components.

B. Preparations of the Chromatographic Column

Chromatographic grade neutral alumina (60 g.) was added to 100 ml. of ethyl acetate and the mixture was stirred gently. The alumina was allowed to settle and excess ethyl acetate was removed by decantation. The remaining alumina-ethyl acetate slurry was dried in a shallow layer for 16 hours at 70° F., after which time the odor of the ethyl acetate was only barely perceptible. The airdried alumina was added to a Kontes Chromaflex No. 25 glass-jacketed chromatographic column containing commercial pentane, which had been cooled and held at 10° C. by circulating cold water through the outer column jacket. The alumina was allowed to settle evenly, aided by occasional gentle tapping of the column. Excess pentane was drained through the column until a "head" of one-sixteenth inch of pentane remained over the flat alumina surface at the top of the column.

C. Separation of Terpenes

The distillate from Part A (23.5 ml.) was carefully layered onto the surface of the alumina column prepared in Part B. The effluent stopcock was opened

slightly and the distillate allowed to percolate into the alumina bed until level with the tope of the alumina adsorbent. Commercial pentane was carefully added to the column without disturbing the surface layer. The effluent stopcock was opened and the pentane allowed to flow freely through the column. Approximately 115 ml. of effluent was obtained before a change in refraction occurred, indicating initial elution of the terpene hydrocarbons. The next 60 ml. of effluent contained all the terpenes. This eluate was collected, and the terpenes were recovered by low-temperature distillation to remove the pentane. Analysis of this fraction indicated that essentially no oxygenated compounds were present.

D. Separation and Recovery of Oxygenated Components

The pentane head on the alumina column (from Part C) was allowed to recede to within one-sixteenth inch of the surface of the alumina and 120 ml. of absolute ethanol was placed carefully on the column. The first 45 ml. of eluate was essentially pure pentane that had occupied the liquid portion of the column prior to addition of the ethanol. A clear change in refraction of the eluate and the nearly concurrent emergence of a pale yellow band signaled the initial elution of the desired oxygenated components. About 75 ml. of ethanol was required to elute the fraction containing the oxygenated components. The eluate was collected and evaporated (to remove the ethanol) on a

Buchi Rotavapor evaporated, yielding 0.6 ml. of terpeneless lemon oil.

A sample of the product was dissolved in ethanol and it was found that this solution was entirely miscible with water. Aqueous solutions for example, of the ethanolic, terpeneless lemon oil retained their typical lemon flavor during storage in green-colored Pyrex, screw-capped (Teflon liners), bottled at ambient temperature for at least one year without loss in flavor or development of off-flavor. In contrast, similar solutions of whole lemon oil developed off-flavors and odors within a few weeks.

Gas chromatographic analysis of the various highly rectified examples showed removal of various amounts of terpenes. As the data collected in Table III demonstrate, the rectification of mint oils such as peppermint oils can be tailored to reduce the amount of those other components which usually are not affected by distillation.

Of the greater interest is the special fractionated peppermint oil containing lower amounts of menthone. This particularly desirable, rectified peppermint oil has very low "head" and "tail" fractions as well as reduced concentrations of other components such as, e.g., 3-octanol, 1-octen-3-ol, menthofuran, and iso-menthone (see Table III).

All percentages throughout the specification are by weight percent of the final composition unless otherwise specified.

TABLE III

COMPARATIVE GAS CHROMATOGRAPHIC ANALYSES OF THE PEPPERMINT OILS

Component	Ex. 1 (%)	Ex. 2 (%)	Ex. 3 (%)	Ex. 4 (%)	Ex. 5 (%)
Isobutyraldehyde	0.04	0	0	0	0
Isovaleraldehyde	0.19	0	0	0	0
A-Pinene	0.73	0	0.10	0	0
B-Pinene	0.95	0	0.25	0	0.02
Sabinene	0.45	0	0.12	0	0.01
Myrcene	0.20	0	0.07	0	0.02
A-Terpinene	0.34	0	0	0	0.07
L-Limonene	1.49	0.11	0.68	0	0.30
1,8-Cineole	4.64	0.37	2.89	0	1.81
Cis-Ocimene	0.30	0.08	0.17	0	0.09
G-Terpinene	0.68	0.20	0.54	0	0.31
P-Cymene	0.18	0	0.15	0	0.05
Terpinolene	0.15	0.14	0.11	0	0.14
3-Octanol	0.24	0.14			
1-Octen-3-Ol	9.17	0.11	0.17	0	0.09
trans-Sabinese	1.04	1.04	0.12	0.18	0.98
Hydrate					
L-Menthone	19.33	23.13	22.28	10.84	23.80
Menthofuran	1.84	2.02	1.95	0.050	2.30
Iso-menthone	2.63	3.30	3.11	2.04	3.16
B-Fourbonene	0.45	0.35	0.43	0.61	0.42
Linalool	0.22	0.23	0.18	0.32	0.33
Menthyl Acetate	5.14	5.15	4.59	7.54	4.42
Neo-menthol	4.04	5.04	5.75	6.48	5.31
B-Caryophyllene	1.62	0.89	1.37	2.07	0.60
Terpinen-4-ol	1.13	1.43	1.62	1.05	1.17

Component	Ex. 1 (%)	Ex. 2 (%)	Ex. 3 (%)	Ex. 4 (%)	Ex. 5 (%)
L-Menthol	41.11	51.35	46.87	63.57	46.77
Pulegone	1.25	1.09	1.48	1.63	1.36
Oermacrene-D	2.12	0.37	1.92	0.70	1.89
Piperitone +	0.99	0.64	0.62	0.92	0.58
Viridiflorol	0.37	0.18	0.10	0	0.18

Ex. 1: Standard peppermint oil
Ex. 2: Highly fractionated and terpeneless oil
Ex. 3: Moderate fractionation
Ex. 4: Special fractionated & lower menthone fraction
Ex. 5: Terpeneless oil, standard deterpenation, "head" fraction cut only.

The following examples serve to provide further appreciation of the invention but are not meant to restrict the effective scope of the claims in any way.

PREPARATION A

For example, the following procedure for Preparation A was undertaken to determine the turbidity readings on the following basic liquid oral composition type mouthrinse product flavored with different types of peppermint oil.

PROCEDURE:

(1) Prepare the mouth wash solution by weighing out peppermint oil (1 gm) andadd 200 ml alcohol.

a. Peppermint Oil Madras, Example #6;

b. Peppermint Oil Blend (Rose Mitchum), Example #7;

c. Peppermint Oil Highly Rectified, Example #8;

(2) To 400 ml. water add the following amounts of Pluronic 407:

Ex. 6A-8A	1.25	g;
Ex. 6B-8B	3	g;
Ex. 6C-8C	6	g;
Ex. 6D-8D	9	g;

(3) Slowly add step (1) to step (2) while mixing rapidly;

(4) Q.S. to 1 liter with water; and

(5) Mix and do turbidity test.

Method of turbidity test:

Nephlometric angle (90°) measurement ratioed to sum of transmitted light and forward scatter light measurements. The results are shown in Table IV.

RESULTS:

TABLE IV: NATIONAL TURBIDITY UNITS

OILS	EX. A	EX. B	EX. C	EX. D
EX. 6	13.8	2.9	2.7	2.7
EX. 7	3.3	2.9	3.1	3.2
EX. 8	2.8	2.2	2.1	2.1

The turbidity measurement are indicate a marked improvement due to the highly rectified peppermint oil, at all concentrations of Pluronic 407 (Example 8A-8D) as shown in Table IV.

PREPARATION II

The solutions presented in Table V were prepared in two parts as follows (as described above):

1) in 500 ml deionized water dissolve the following ingredients:
   Glycerin, sodium saccharin, citric acid, sodium cetrate, sodium lauryl sulfate, yellow #10, green #3, poloxamer 407 (Part 1).
2) Dissolve the following ingredients in Ethanol: spearmint, methyl salicylate, menthol, and peppermint (Part 2).

TABLE V:
PEPPERMINT COMPOSITION

INGREDIENTS		A	B	C	D	E	G	G	H
Ethanol 95%	ml	150	->	->	->	->	->	->	->
Spearmint Oil	g	0.3	->	->	->	->	->	->	->
Methyl Salicylate	g	0.45	->	->	->	->	->	->	->
Menthol	g	0.4	->	->	->	->	->	->	->
Glycerin	g	70	->	->	->	->	->	->	->
Sodium Saccharin	g	0.7	->	->	->	->	->	->	->
Citric Acid	g	0.094	->	->	->	->	->	->	->
Sodium Citrate	g	0.783	->	->	->	->	->	->	->
Sodium Lauryl Sulfate	g	0.75	->	->	->	->	->	->	->
D&C Yellow #10	g	0.010	->	->	->	->	->	->	->
FD&C Green #3	g	0.0025	->	->	->	->	->	->	->
Poloxamer 407	g	6	5	4	3	6	5	4	3
Rose Mitcham Peppermint	g	1.0	1.0	1.0	1.0	-	-	-	-
Far West Terpene-less Peppermint	g	-	-	-	-	1.0	1.0	1.0	1.0
Water g s to	l	1.0	->	->	->	->	->	->	->
Turbidity (NTU)		6.5	8.3	7.0	8.4	2.0	2.0	2.1	2.2

(->) represents ditto quantity; referring back to column "A".

3) Add part 2 to part 1 while mixing rapidly; and

4) Adjust to volume with water.

The experimental results in terms of turbidity (NTU) describe the comparative advantage of the Farwest terpeneless peppermint oil which effects products of superior clarity in solution, i.e., lower turbidity, thus reducing the need for surfactants, which add cost

adversely affect taste, and may bind ingredients such as may be present in the composition of a mouthwash (See TABLE V) and may shorten shelf-life.

Consequently, an organoleptic test the comparison test of Table V determined that a mouthwash in accordance with the present invention having better clarity and lower surfactant level also resulted in a cleaner freshly intense minty taste.

As would be understood by those skilled in the art, many processing parameters, flavoring agents and other embodiments of the present invention can be varied or changed to a minor degree. However, these minor modifications should not be regarded as a departure from the invention as described and defined by the following claims.

WHAT IS CLAIMED IS:

1. A liquid oral hygiene composition with an improved minty flavor comprising:
   (a) a flavor oil substantially free of undesirable components;
   (b) one or more of a surfactant component ranging by weight up to about 5%; and
   (c) not more than about 30% by weight ethyl alcohol.
2. The liquid oral hygiene composition of claim 1, wherein the undesirable components comprising terpenes have been removed from the flavor oil by extraction or fractionation.
3. The liquid oral hygiene composition of claim 2, wherein the terpenes are characterized by fractions of monoterpenes and sesquiterpenes.
4. The liquid oral hygiene composition of claim 1 or 2, wherein the flavor oil is a mint oil.
5. The liquid oral hygiene composition of claim 3, wherein the mint oil comprises peppermint, spearmint, corn mint and mixtures thereof.
6. The liquid oral hygiene composition of claim 2 or 5, wherein the mint oil is present in concentrations ranging from about 0.01% to about 5.0% by weight of the composition.
7. The liquid oral hygiene composition of claim 1, further comprising an intense sweetening agent the concentration ranging up to about 3.0% by weight of the composition.

8. The liquid oral hygiene composition of claim 1, wherein the intense sweetening agent is selected from the group consisting of sodium or calcium saccharin salts, the free acid form of saccharin, cyclamate salts, chlorosucrose derivatives, dipeptide compounds, acesulfame-K, and blends thereof.

9. An improved mint flavored clear mouthwash composition comprising substantially terpene free peppermint oil; one or more surfactant components ranging from about 0.01% to about 3% by weight; ethyl alcohol ranging up to about 35% by weight; a sweetening agent ranging up to 3.0% by weight; an antimicrobial quaternary ammonium salt ranging from about 0.005% to about 5%; a fluorine providing compound ranging from about 0.001% to about 0.1%; a coloring agent; and a pharmaceutically acceptable buffer ranging from about 0.05% to about 0.5% by weight.

INTERNATIONAL SEARCH REPORT

Interr nal Application No
PCT/US 94/05118

A. CLASSIFICATION OF SUBJECT MATTER
IPC 5 A61K7/26 A61K7/16

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 5 A61K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO,A,93 08792 (WARNER-LAMBERT COMPANY) 13 May 1993 see the whole document ---	1-9
X	EP,A,0 310 299 (PROCTER & GAMBLE) 5 April 1989 see example 2 ---	1-9
X	WO,A,93 04664 (7-L CORPORATION) 18 March 1993 see claims 1,4,6-9 --- -/--	1-9

[X] Further documents are listed in the continuation of box C.

[X] Patent family members are listed in annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
27 September 1994	10. 10. 94

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax (+31-70) 340-3016	Fischer, J.P.

Form PCT/ISA/210 (second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

Inter nal Application No
PCT/US 94/05118

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	JANISTYN 'HANDBUCH DER KOSMETIKA UND RIECHSTOFFE, BAND 2 : DIE KÖRPERPFLEGEMITTEL' 1973 , 2.AUFL., HÜTHIG , HEIDELBERG, DE see page 789, Mundwasser emulgiert; page 793, Zahnreinigungsmittel flüssig; page 793, Zahnreinigungslotion; page 794, Zahnputzmittel flüssig	1-9

Form PCT/ISA/210 (continuation of second sheet) (July 1992)

INTERNATIONAL SEARCH REPORT

information on patent family members

Inte nal Application No
PCT/US 94/05118

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
WO-A-9308792	13-05-93	US-A-	5298238	29-03-94
		AU-A-	2775392	07-06-93
EP-A-0310299	05-04-89	NONE		
WO-A-9304664	18-03-93	US-A-	5174990	29-12-92
		AU-A-	2578892	05-04-93
		US-A-	5310546	10-05-94